36691X

Journal of Exposure Analysis and Environmental Epidemiology (1999) 9, 381–392

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National Human Exposure Assessment Survey (NHEXAS): distributions and associations of lead, arsenic and volatile organic compounds in EPA Region 5

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Clayton, CA et al 1999A

The National Human Exposure Assessment Survey (NHEXAS) Phase I field study conducted in EPA Region 5 provides extensive exposure data on approximately 250 study participants selected via probability sampling. Associated environmental media and biomarker (blood, urine) concentration data were also obtained to aid in the understanding of relationships of the exposures to both contaminant sources and doses. Distributional parameters for arsenic (As), lead (Pb), and four volatile organic compounds (VOCs)—benzene, chloroform, tetrachloroethylene, and trichloroethylene—were estimated for each of the relevant media using weighted data analysis techniques. Inter-media associations were investigated through correlation analysis, and longitudinal correlations and models were used to investigate longitudinal patterns. Solid food appeared to be a major contributor to urine As levels, while Pb levels in household (HH) dust, personal air, and beverages all were significantly associated with blood Pb levels. Relatively high (>0.50) longitudinal correlations were observed for tap water Pb and As, as compared to only moderate longitudinal correlations for the personal air VOCs.

Introduction

The National Human Exposure Assessment Survey (NHEXAS) of the U.S. Environmental Protection Agency (EPA) is a multi-contaminant, multi-media, three-phase program; the program is described in Pellizzari et al. (1995). The present paper is concerned with one of the NHEXAS Phase I field studies. This study was conducted in EPA's Region 5 (six States in the Great Lakes Region) by a consortium of institutions, researchers, and consultants led jointly by the Research Triangle Institute (RTI) and the Environmental and Occupational Health Sciences Institute (EOHSI). The aims of this first phase were to field test survey and measurement methodology for Phase II, to collect data useful for addressing regional environmental problems, and to collect information on cost components (survey, monitoring, analysis) and variance components for optimizing the Phase II design. The Region 5 study focused on six primary contaminants: arsenic (As), lead (Pb), and four volatile organic compounds (VOCs) — benzene, chloroform, tetrachloroethylene (PERC), and trichloroethylene (TCE). Media sampled included house dust, air (6-day personal, indoor, and outdoor integrated samples), tap water, and food and beverages (4-day composite samples); urine and blood served as the main biomarkers. Questionnaires administered to participants elicited both general demographic and behavioral data, as well as specific activity and microenvironmental data for the 6-day monitoring periods. Details on the study objectives, design, and methods can be found in Pellizzari et al. (1995).

The overall goals of this paper are (a) to provide information on the target population's distributions of exposure levels of the primary NHEXAS contaminants; (b) to provide similar distributional information for environmental media and for biomarkers; (c) to provide insight into associations of the environmental measures, the exposure measures, and the biomarkers; and (d) to explore longitudinal patterns in the exposure data. The results should furnish a useful background for subsequent investigation of the data. In particular, the results should help focus future data analyses and model building efforts on those exposure pathways found to be most important.

1. Abbreviations: As, arsenic; CL, confidence limit; corr, correlation; CV, coefficient of variation; EOHSI, Environmental and Occupational Health Sciences Institute; EPA, U.S. Environmental Protection Agency; GSD, geometric standard deviation; HH, household; NHEXAS, National Human Exposure Assessment Survey; LOD, limit of detection; Pb, lead; PERC, tetrachloroethylene; PSU, primary sampling unit; RTI, Research Triangle Institute; TCE, trichloroethylene; VOC, volatile organic compound.

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Revised 30 September 1998.

Methods

Study Design

The target population for the Region 5 study consisted of all noninstitutionalized persons residing in households (HHs) in six States (Illinois, Indiana, Ohio, Michigan, Minnesota, Wisconsin) during the time of data collection (July 1995–May 1997). The sampling design utilized for the Region 5 study was a stratified, four-stage probability sampling design. The first-stage sample involved 32 primary sampling units (PSUs) (usually a county), selected with probabilities proportional to 1990 Census counts of occupied housing units; these PSUs were grouped into four analysis strata (temporal categories indicating when data collection was scheduled). Three or four sample areas (area segments defined by Census blocks) were then selected within each sample county as second-stage units. A simple random sample of eight housing units (the third-stage units) was then fielded in each segment. Participants were then selected from HH rosters (at the fourth stage); the goal was to acquire approximately nine participants in each PSU. A subsample of participants was also asked to enroll in the longitudinal component of the study (i.e., participation in two additional 6-day monitoring periods). Details on the study design and its rationale are given in Pellizzari et al. (1995).

Data Collection

The study involved collection of both questionnaire data and physical measurements data; the data were acquired at the following five stages.

(1) Administration of a Descriptive Questionnaire to selected HHs in the six-State region — to acquire basic HH demographic data and to provide data (e.g., number of HH members) for selecting study participants.

(2) Administration of a Baseline Questionnaire to a selected person in a subset of those HHs — to acquire general HH and participant characteristics data (e.g., use of heating/cooling systems, tobacco use, occupation).

(3) Collection of physical samples and questionnaire data for study participants (a subset of the Baseline respondents) relating to the primary monitoring period (Visit 1) (questionnaire data dealing with participants' activities, microenvironments, dietary patterns, etc.).

(4) Collection of physical samples and questionnaire data for a subset of the Visit-1 participants during a follow-up period (Visit 2).

(5) Collection of physical samples and questionnaire data for a subset of the Visit-1 participants during a second follow-up period (Visit 3).

The basic unit of observation was a HH or person for the first two types and was a person–period or HH–period for the latter three. The two longitudinal monitoring periods were conducted by the participants utilizing kits sent out by mail — i.e., the terminology "Visit" is used simply to distinguish these different monitoring periods.

As noted above, this paper focuses on Pb and As and on four VOCs and on those media for which data were adequate to generate population estimates. These media are listed in Table 1, along with the number of visits per participant for which such data were obtained. Sample collection and analysis methods have been described elsewhere (Pellizzari et al., 1995; Thomas et al., 1998).

Statistical Analysis

Because the sampling design is not a simple random sample, and because not all participants were selected with equal probabilities, weighted data analyses are appropriate when possible. Since some participants did not provide data for some of the media, different sampling weights were developed for each environmental medium and chemical type (metals and particles, or VOCs). In addition, since subsamples of the participants were used for Visits 2 and 3, separate weights were developed for each visit. These visit-specific weights are appropriate when estimates based on a single visit are desired and when no adjustment for time of sampling is desired. To generate estimates based on all data from all available visits, two types of combined sampling weights were generated — one for which no adjustment for time of sampling was used, and one (referred to as annualized weights) for which such an adjustment was employed. The annualized weights were employed in most

Table 1. Types of data.

Medium	Unit of observation	Metals	VOCs
Personal air	Person–Period	1 visit[a]	up to 3 visits
Indoor air	HH–Period	1 visit[a]	up to 3 visits
Outdoor air	HH–Period	1 visit[a]	1 visit
House dust			
Surface wipe	HH–Period	1 visit	
Window sill wipe	HH–Period	1 visit	
Water			
Drinking	HH–Period		1 visit
Standing tap	HH–Period	up to 3 visits	
Flushed tap	HH–Period	up to 3 visits	
Solid foods	Person–Period	1 visit	–
Beverages	Person–Period	1 visit	
Blood	Person–Period	1 visit (Pb only)	1 visit
Urine			
Day 7 of monitoring period	Person–Period	1 visit (As only)	
Average of days 3 and 7	Person–Period	1 visit (As only)	

[a]From particle samples (particles with diameter of approximately 50 μm or less).

of the analyses described herein. The initial weights for the sample and subsamples were adjusted for nonresponse using weighting-class adjustment procedures, longitudinal models for propensity to respond, and exponential, generalized ranking models (Folsom, 1991; Kalton and Maligalig, 1991).

Population estimates of means, medians, and other percentiles were generated using weighted data analysis techniques. For instance, a population mean for a contaminant concentration would be estimated as $\sum wy/\sum w$, where w is the sampling weight and y is the concentration, and where the summation is over those participants providing data on y. Since all of the reported summary statistics (e.g., percentage measurable, mean, median, 90th percentile) were computed using the sampling weights, the estimates apply to the Region 5 target population. Confidence intervals for such population parameters were generated using SUDAAN software, in order to properly account for the sampling design in computing variances of the estimates (Shah et al., 1997). For percentiles, SUDAAN determines the confidence intervals by inverse interpolation of the weighted empirical distribution function (i.e., confidence intervals for proportions are first estimated and then

Table 2. Estimated parameters for Region 5: As.

Medium	N	Percentage measurable	Mean	50th (median)	90th	Percentage measurable CL	Mean CL	Median CL	90th CL
Personal air (ng/m^3)	169	94.1	1.56	0.90	3.29	88.9	1.13	0.67	2.03
						99.2	2.00	1.13	4.94
Indoor air (ng/m^3)	218	84.9	0.65	0.49	1.15	75.8	0.50	0.39	0.89
						94.0	0.80	0.59	1.54
Outdoor air (ng/m^3)	85	91.1	0.74	0.71	1.14	78.4	0.60	0.58	0.98
						100.0	0.89	0.80	1.25
Surface dust (ng/cm^2)	247	24.4	0.47	0.26	1.01	17.7	0.35	0.20	0.74
						31.1	0.59	0.34	1.27
Surface dust (μg/g)	245	24.6	17.19	5.43	34.74	17.9	10.78	4.43	28.23
						31.4	23.60	7.38	48.07
Window sill dust (ng/cm^2)	240	37.7	0.78	0.34	1.33	28.2	0.43	0.25	1.00
						47.2	1.13	0.48	1.66
Window sill dust (μg/g)	240	37.7	10.43	5.30	22.42	28.2	8.33	4.35	18.49
						47.2	12.52	6.65	29.53
Standing tap water (μg/l)	443	96.5	1.12	0.72	1.68	93.6	0.71	0.63	1.19
						99.3	1.52	0.88	2.14
Flushed tap water (μg/l)	445	96.5	1.12	0.73	1.59	93.5	0.72	0.63	1.20
						99.5	1.52	0.87	2.09
Solid food (μg/kg)	159	99.7	17.21	7.72	43.05	99.1	12.26	6.18	23.66
						100.0	22.16	9.86	46.36
Beverages (μg/kg)	160	90.4	1.47	0.88	3.30	83.9	1.17	0.80	2.27
						96.8	1.77	1.01	3.64
Food+Beverages (μg/kg)	156	99.7	6.31	3.48	14.70	99.0	4.74	2.55	8.96
						100.0	7.87	4.69	18.59
Food intake (μg/day)	159	99.7	12.64	4.99	31.63	99.1	8.51	3.70	18.71
						100.0	16.78	7.61	44.68
Beverage intake (μg/day)	160	90.4	2.26	1.30	4.50	83.9	1.73	1.18	3.83
						96.8	2.80	1.50	5.49
Food+Beverage intake (μg/day)	156	99.7	15.10	7.18	35.36	99.0	10.77	5.16	23.35
						100.0	19.43	9.99	49.16
Urine (μg/l)–average	202	58.9	29.32	3.65	30.54	46.9	−1.01	[a]	15.17
						70.8	59.66	7.78	46.29
Urine (μg/l)–day 7	197	48.7	33.59	[a]	26.43	34.8	1.14	[a]	13.30
						62.6	66.04	5.47	43.31

[a]Indicates that quantity could not be estimated.

Estimates for indoor and outdoor air and dust and water apply to the target population of Region 5 HHs; estimates for other media apply to the target population of Region 5 residents.

translated into the scale of the parameters). This results in intervals that are not necessarily symmetrical about the estimated percentile.

To investigate associations among media, several types of correlations can be used — e.g., Pearson correlations, Spearman (i.e., rank) correlations, and Pearson correlations of log-transformed data. The original-scale Pearson correlations can be particularly sensitive to extreme values. Unlike the Spearman correlations, the Pearson correlations can be (and were) weighted. The log-transformed correlations are less sensitive to extreme high values, but can be sensitive to small values (e.g., numeric values assigned to measurements below limits of detection (LODs)). The Spearman correlations are invariant to changes in scale, since they are based on ranks; the fact that they are not sensitive to a few extreme values sometimes causes them to be overly conservative (in the sense of not revealing an association when one actually exists).

To examine longitudinal patterns (for those media and analytes with such data), pairs of observations for the same participant were grouped by the amount of time between observations (from 1 to 8 months); correlations of the log-scale measurements were then computed for each such group. Since there are only a small number of observations available for computing these correlations, they are subject to wide variation. Hence, the resultant set of eight correlations were smoothed using an exponential type of model that allows the correlations to decline over time (a limiting form of the model allows the correlations to remain constant over time). A model-based approach for estimating

Table 3. Estimated parameters for Region 5: Pb.

Medium	N	Percentage measurable	Mean	50th (median)	90th	Percentage measurable CL	Mean CL	Median CL	90th CL
Personal air (ng/m^3)	167	81.6	26.83	13.01	57.20	71.3	17.60	11.13	31.18
						92.0	36.06	18.13	85.10
Indoor air (ng/m^3)	213	49.8	14.37	6.61	18.50	37.2	8.76	4.99	12.69
						62.3	19.98	8.15	30.31
Outdoor air (ng/m^3)	87	73.8	11.32	8.50	20.36	56.3	8.16	7.14	12.60
						91.3	14.47	10.35	34.91
Surface dust (ng/cm^2)	245	92.1	514.43	5.96	84.23	87.4	−336.6	3.37	26.52
						96.8	1365.5	10.94	442.63
Surface dust (μg/g)	244	92.1	463.09	120.12	698.92	87.4	188.15	83.85	411.84
						96.8	738.04	160.59	1062.8
Window sill dust (ng/cm^2)	239	95.8	1822.6	16.76	439.73	92.5	481.49	10.44	106.34
						99.0	3163.6	39.41	4436.2
Window sill dust (μg/g)	239	95.8	954.07	191.43	1842.8	92.5	506.70	140.48	1151.3
						99.0	1401.4	256.65	2782.5
Standing tap water (μg/l)	444	98.8	3.92	1.92	9.34	97.6	3.06	1.49	7.87
						100.0	4.79	2.74	12.35
Flushed tap water (μg/l)	443	78.7	0.84	0.33	1.85	70.7	0.60	0.23	1.21
						86.7	1.07	0.49	3.04
Solid food (μg/kg)	159	100.0	10.47	6.88	14.88	100.0	6.87	6.44	10.78
						100.0	14.07	8.04	19.08
Beverages (μg/kg)	160	91.5	1.42	0.99	2.47	85.2	1.13	0.84	2.06
						97.8	1.72	1.21	3.59
Food+Beverages (μg/kg)	156	100.0	4.48	3.10	6.37	100.0	2.94	2.66	4.89
						100.0	6.02	3.52	8.00
Food intake (μg/day)	159	100.0	7.96	4.56	12.61	100.0	4.25	3.68	9.27
						100.0	11.68	5.36	16.38
Beverage intake (μg/day)	160	91.5	2.15	1.41	4.45	85.2	1.66	1.18	3.15
						97.8	2.64	1.60	5.65
Food+Beverage intake (μg/day)	156	100.0	10.20	6.40	16.05	100.0	6.52	5.21	13.31
						100.0	13.89	7.78	18.85
Blood (μg/dl)	165	94.2	2.18	1.61	4.05	88.2	1.78	1.41	3.24
						100.0	2.58	2.17	5.18

Estimates for indoor and outdoor air dust and water apply to the target population of Region 5 HHs; estimates for other media apply to the target population of Region 5 residents.

the distribution of long-term exposures (or concentrations) was also attempted. The method, described later, is an adaptation of the method of Wallace et al. (1994) that relies on an assumption of lognormality for the population distribution of exposures for any averaging time.

Results and discussion

As and Pb

Table 2 provides pertinent summary statistics for As for various media. The table gives the number of observations (*N*), which is the number of person–periods or HH–periods for which usable data were obtained. The statistics include the percentage measurable (i.e., the percentage of the target population of persons or HHs estimated to have As levels above the LOD) and estimates of the target population's mean, median (50th percentile), and 90th percentile. Confidence intervals for these parameters are also shown: the first row for a given medium gives the lower 95% confidence limit (CL), and the second row gives the upper 95% CL. Except for outdoor air, the means are generally 1.5 to 3 times larger than the medians, indicating that the As distributions are highly skewed to the right. The urine distributions are the most highly skewed; the mean for the day 7 measurement, for instance, actually is above the 90th percentile. Several interesting properties can be noted.

Personal air levels for As tend to be higher than either indoor or outdoor levels; this is at least partly due to a similar pattern (not shown in the table) of exposures and concentrations for particles (e.g., median particle exposure of 101 $\mu g/m^3$ versus median indoor concentration of 34.4 $\mu g/m^3$), a result consistent with results reported in prior studies (e.g., Clayton et al., 1993).

Distributions for standing and flushed tap water concentrations appear to be very similar.

Levels of As in solid foods are much higher than those in beverages.

As levels in urine are not detectable for about 50% of the population; some individuals, however, have very high levels as compared to most other individuals.

Table 3 provides comparable estimates for Pb.

Personal air levels for Pb, like As, tend to be higher than either indoor or outdoor levels.

Window sill levels are higher than the main-living-area surface levels, suggesting Pb-based paint (especially in older homes) and/or infiltration of outdoor particles as potential sources.

Distributions for the standing and flushed tap water concentrations are quite different, with standing tap water exhibiting much higher levels; some HHs, probably those

Table 4. Associations of biological markers with environmental concentration and exposure measures–As and Pb.

Medium	As levels						Pb levels		
	Corr (medium, avg. urine)			Corr (medium, day 7 urine)			Corr (medium, blood)		
	N	Pearson[a]	Spearman	*N*	Pearson[a]	Spearman	*N*	Pearson[a]	Spearman
Personal air	150			146			126	0.13	0.17
Indoor air	190			185			153		0.16*
Outdoor air	78			75			66		
Surface dust–load	200			195			164		0.25**
Surface dust–concentration	198		0.11	193		0.09	163	0.14	0.17*
Window sill dust–load	197		−0.08	192		−0.08	160	0.12	0.16*
Window sill dust–concentration	197			192		0.10	160	0.25**	0.10
Standing tap water	199	0.10		194	0.12		163		0.09
Flushed tap water	201			196			164	0.21**	0.19*
Solid food–concentration	149	0.10		147	0.17*	0.08	130		
Beverages–concentration	149	−0.10		147			130	0.20*	
Food+Beverages–concentration	145		0.09	143		0.11	127	0.18*	0.08
Solid food–intake (μg/day)	149			147	0.10	0.09	130		
Beverages–intake (μg/day)	149			147			130	0.21*	
Food+Beverages–intake (μg/day)	145			143	0.08		127	0.16	
Solid food–intake (μg/kg body weight/day)	146			144	0.10	0.10	127	0.08	
Beverages–intake (μg/kg body weight/day)	146			144			127	0.23**	
Food+Beverages–intake (μg/kg body weight/day)	142			140			124	0.19*	

*Statistically significant at the 0.05 level.
**Statistically significant at the 0.01 level.
[a]Pearson correlations are weighted to reflect the target population of HHs or persons.
Correlations less than 0.08, in absolute value, are omitted.

Table 5. Spearman correlations among media for As.

Medium	Indoor air		Outdoor air		Surface dust		Standing water		Flushed water		Solid food		Beverages		Food+Beverages	
	n	Corr	*n*	Corr	*n*	Corr	*n*	Corr	*n*	Corr	*n*	Corr	*n*	Corr	*n*	Corr
Personal air	166	0.64**	76	0.26*	168	0.23**	166	0.23**	169	0.25**	132	0.01	133	0.01	129	0.07
Indoor air			84	0.56**	217	0.28**	215	0.15*	218	0.19**	149	0.09	150	0.02	146	0.08
Outdoor air					84	0.06	85	−0.03	85	0.03	70	0.04	70	0.18	67	0.04
Surface dust							244	0.14*	247	0.12	158	−0.00	159	−0.12	155	−0.04
Standing water									442	0.94**	157	0.08	158	0.15	154	0.12
Flushed water											159	0.05	160	0.16*	156	0.10
Solid food													159	0.18*	155	0.89**
Beverages															156	0.41**

*Statistically significant at the 0.05 level.
**Statistically significant at the 0.01 level.
n=number of paired observations (e.g., person−periods).

with pipes having Pb-based solder, exhibit high standing-water Pb levels.

Levels of Pb in solid foods are higher than those in beverages.

Pb levels in blood are detectable for about 94% of the population; most individuals have levels well below 10 μg/dl (Action level for children, Centers for Disease Control and Prevention).

Table 4 furnishes insight into the association of the exposures and media concentrations with the biomarkers. For As, it presents the following.

- The number of participants having data for the given medium (as identified by the row) and for urine. (For most participants, the urine value is the average of measurements made on days 3 and 7 of the monitoring period; for a few participants, only one of the two urine values was available and its concentration value was used. Participants' food samples were collected between days 3 and 7 of their monitoring period).
- The weighted Pearson correlation between the exposures or concentrations or intakes associated with the given medium and the average urine levels (sampling weights were based on those associated with the urine data).
- The Spearman (rank) correlation between the exposures or concentrations or intakes associated with the given medium and the average urine levels.
- The same three statistics for the day-7 urine samples.

The results suggest that there is very little association between the As levels in average urine and those measured in the environment; for the day-7 urine (which occurred after the food and beverage collection period), some association with food levels (primarily solid food) was evident. For Pb, Table 4 gives similar statistics (in the last three columns) with blood rather than urine as the biomarker. Blood Pb levels appear to be positively correlated with the Pb levels that occur in (personal and indoor) air, dust, flushed water, and beverages.

Tables 5 and 6 give inter-media Spearman correlations for As and Pb, respectively, and indicate the following.

For As, personal air exposures exhibit a strong association with indoor air concentrations (corr=0.64), and indoor air

Table 6. Spearman correlations among media for Pb.

Medium	Indoor air		Outdoor air		Surface dust		Standing water		Flushed water		Solid food		Beverages		Food+Beverages	
	n	Corr	*n*	Corr	*n*	Corr	*n*	Corr	*n*	Corr	*n*	Corr	*n*	Corr	*n*	Corr
Personal air	161	0.63**	75	0.25*	166	0.30**	166	0.20**	166	0.27**	131	0.14	132	0.22*	128	0.11
Indoor air			84	0.47**	211	0.22**	211	0.10	212	0.23**	146	0.10	147	0.08	144	0.07
Outdoor air					86	0.25*	87	0.15	86	0.17	72	0.09	72	0.23*	69	0.12
Surface dust							243	0.09	244	0.18**	157	0.11	158	0.18*	154	0.13
Standing water									441	0.64**	157	0.14	158	0.46**	154	0.33**
Flushed water											158	0.12	159	0.40**	155	0.24**
Solid food													159	0.04	155	0.63**
Beverages															156	0.58**

*Statistically significant at the 0.05 level.
**Statistically significant at the 0.01 level.
n=number of paired observations (e.g., person−periods).

concentrations exhibit a strong association with outdoor air concentrations (corr=0.56); otherwise, the air correlations are generally weak (<0.30), though some are statistically significant (e.g., dust with personal and indoor air).

Flushed water As concentrations are very highly correlated (corr=0.94) with standing water concentrations, as might be anticipated, and are also weakly (but significantly) correlated with personal and indoor air levels and with beverage concentrations.

Pb exhibits similar patterns, except that the standing versus flushed water association is not nearly so strong (corr=0.64) and the water versus beverage correlations are stronger (0.46 and 0.40 for standing and flushed water, respectively).

Based on the strength of associations, the contribution to the dietary pathway for As appears to come principally from the solid foods, whereas beverages appear to be the primary contributor for Pb.

Table 7. Estimated parameters for Region 5: VOCs.

Compound	Medium	*N*	Percentage measurable	Mean	50th (median)	90th	Percentage measurable CL	Mean CL	Median CL	90th CL
Benzene	Personal air ($\mu g/m^3$)	386	99.7	7.52	5.37	13.71	99.1	6.30	4.83	10.15
							100.0	8.74	5.55	17.10
	Indoor air ($\mu g/m^3$)	402	99.8	7.21	4.35	12.95	99.5	4.65	4.02	8.71
							100.0	9.78	4.99	16.85
	Outdoor air ($\mu g/m^3$)	97	100.0	3.61	2.92	5.62	100.0	3.04	2.62	4.72
							100.0	4.18	3.79	5.98
	Drinking water ($\mu g/l$)	247	5.9	X	X	X	X	X	X	X
	Blood ($\mu g/l$)	143	91.3	0.21	0.14	0.44	86.5	0.17	0.10	0.36
							96.1	0.26	0.19	0.54
Chloroform	Personal air ($\mu g/m^3$)	384	68.3	2.34	1.96	4.54	57.4	1.88	1.41	3.91
							79.2	2.81	2.42	6.19
	Indoor air ($\mu g/m^3$)	402	72.8	2.64	2.13	6.24	61.9	2.13	1.49	4.21
							83.8	3.14	2.59	7.07
	Outdoor air ($\mu g/m^3$)	97	48.2	1.57	0.86	4.87	37.0	1.09	0.38	2.18
							59.4	2.06	2.07	4.94
	Drinking water ($\mu g/l$)	247	80.7	15.19	5.15	47.04	68.3	7.81	1.05	23.21
							93.0	22.57	14.90	56.16
	Blood ($\mu g/l$)	125	40.9	0.07	0.02	0.10	26.7	0.02	0.02	0.06
							55.0	0.12	0.03	0.21
PERC	Personal air ($\mu g/m^3$)	386	61.3	31.92	1.98	10.78	49.5	−15.17	1.50	7.41
							73.0	79.02	2.96	18.77
	Indoor air ($\mu g/m^3$)	402	57.1	5.82	1.89	6.83	44.0	2.45	1.21	5.20
							70.2	9.19	2.70	7.55
	Outdoor air ($\mu g/m^3$)	97	50.0	2.49	1.24	6.71	37.1	1.63	0.63	3.69
							62.9	3.36	2.68	7.16
	Drinking water ($\mu g/l$)	247	6.9	X	X	X	X	X	X	X
	Blood ($\mu g/l$)	147	38.7	0.21	0.05	0.16	28.0	0.07	0.05	0.12
							49.5	0.35	0.05	0.66
TCE	Personal air ($\mu g/m^3$)	386	39.4	5.27	0.63	5.98	28.1	−0.08	0.44	2.70
							50.8	10.62	0.97	26.19
	Indoor air ($\mu g/m^3$)	402	36.1	3.84	0.56	2.28	25.6	−0.13	0.32	1.80
							46.5	7.81	0.84	3.29
	Outdoor air ($\mu g/m^3$)	97	26.4	1.11	0.32	1.57	12.9	0.06	0.13	1.00
							40.0	2.16	0.64	1.98
	Drinking water ($\mu g/l$)	247	8.1	X	X	X	X	X	X	X
	Blood ($\mu g/l$)	149	7.0	X	X	X	X	X	X	X

Estimates for indoor and outdoor air and for water apply to the target population of Region 5 HHs; estimates for personal air and blood apply to the target population of Region 5 residents. (X=statistic not reported due to low percentage measurable).

VOCs

Table 7 presents the weighted summary statistics characterizing the distributions of Region 5 person–periods (personal air and blood) or HH–periods (indoor air, outdoor air, drinking water). These results show the following.

All of the distributions are skewed to the right, with means generally about 1.5 to 3 times higher than the median of the distribution. PERC and TCE are exceptions, mainly for personal air, where the mean was inflated due to the presence of a few extremely large values (perhaps evidence of occupational exposure).

Benzene, PERC, and TCE personal air levels tend to exceed both indoor and outdoor levels. This pattern is consistent with a number of prior studies (e.g., see Wallace, 1989).

Except for chloroform, most drinking water levels were below detectable levels, as were the blood TCE values.

Table 8 presents correlations of the VOC environmental concentrations and exposures with the biomarker levels. Statistically significant correlations with the biomarkers were generally observed for both personal air and indoor air (TCE indoor air was an exception).

Table 9 shows the inter-media rank (Spearman) correlations for each of the four VOCs. For personal air exposures, strong positive correlations (>0.50) with indoor air occur for all four compounds and strong positive correlations with outdoor air are evident for three of the four. A weaker personal versus outdoor air correlation was observed for benzene (0.26), probably due to the strong influence of tobacco smoking on personal and indoor air benzene levels. (Spearman correlations of personal air benzene levels with many of the smoking-related activity variables reported by participants, such as the amount of time in the presence of smokers, or the number of cigarettes smoked, were frequently over 0.50. Rarely were any other correlations of activity variables with exposure data above 0.30.) For chloroform, positive associations were observed between water levels and personal (and indoor) levels; TCE exhibited a negative association between the water levels and the air levels. Water and air levels were not significantly correlated for benzene and PERC.

Table 9. Spearman correlations of exposure and environmental media concentrations: VOCs.

Compound	Medium	Indoor air		Outdoor air		Drinking water	
		N	Corr	*N*	Corr	*N*	Corr
Benzene	Personal air	243	0.62**	95	0.26*	242	−0.07
	Indoor air			97	0.10	246	0.06
	Outdoor air					96	−0.29
Chloroform	Personal air	242	0.59**	94	0.53**	241	0.38**
	Indoor air			97	0.32**	246	0.26**
	Outdoor air					96	0.06
PERC	Personal air	243	0.65**	95	0.64**	242	−0.06
	Indoor air			97	0.48**	246	−0.09
	Outdoor air					96	−0.11
TCE	Personal air	243	0.55**	95	0.75**	242	−0.20**
	Indoor air			97	0.44**	246	−0.20**
	Outdoor air					96	−0.38**

*Statistically significant at the 0.05 level.
**Statistically significant at the 0.01 level.
N=number of paired observations.

Table 8. Associations of blood levels with environmental concentration and exposure measures—VOCs.

Compound	Medium	Corr (medium, blood)[a]		
		N	Pearson[b]	Spearman
Benzene	Personal air	140	0.52**	0.31**
	Indoor air	143	0.36**	0.37**
	Outdoor air	59	0.38**	−0.14
	Drinking water	143		
Chloroform	Personal air	123	(I)	0.09
	Indoor air	125	(I)	0.09
	Outdoor air	52	(I)	−0.13
	Drinking water	125	(I)	0.10
PERC	Personal air	144	0.95*	0.37**
	Indoor air	147	0.33**	0.17*
	Outdoor air	63		
	Drinking water	147	0.12	
TCE	Personal air	146	0.94**	0.17*
	Indoor air	149		0.18*
	Outdoor air	63		0.17
	Drinking water	149		0.13

*Statistically significant at the 0.05 level.
**Statistically significant at the 0.01 level.
[a]Correlations less than 0.08, in absolute value, are omitted.
[b]Pearson correlations are weighted to reflect the target population of HHs or persons.
(I)=incomplete data, weighted results not reported.

Longitudinal Patterns

As noted in Table 1, longitudinal data were available for some media/analyte combinations (e.g., water metals and personal air VOCs). Approximately 100 participants had a second visit and approximately 90 had a third visit; data from these visits can thus be paired to form about 280 pairs (about 100 first/second pairs, about 90 first/third pairs, and about 90 second/third pairs). These pairs can then be grouped according to their time differences. These differences, when expressed in months (the number of days divided by 30) generally fell, by design, between 1 and 8 months, inclusive. Correlations between the pairs can then be computed, by lag time, to determine the extent to which participants' exposures (or HH's concentrations) tend to fluctuate over time. High lag correlations indicate that individuals who have high (low) levels relative to most others will tend to continue to have high (low) levels in subsequent visits. Low correlations, on the other hand, would tend to indicate a larger intra-person variation relative to the inter-person variation.

Table 10 presents these correlations, along with the relevant sample sizes. The correlations were computed for the logarithms of the concentrations in order to ameliorate the impacts of extreme values. The correlations are based on small sample sizes and are therefore subject to substantial variation. To compensate for this, and to provide an overall indication of the pattern of the correlations across time, a model was used to smooth the directly computed correlations (denoted as L. Corr); the smoothed estimates, which are denoted as "Est." in the table, are the results of fitting a weighted nonlinear least-squares model to the correlations. The weights used were the sample sizes, and the model is as shown in the footnote to Table 10. The model allows for the correlations to decay exponentially or to remain constant over time. The standing and flushed water results for As are very similar: very high lag 1 correlations that tend to gradually decline with time. The Pb standing-water results exhibit a fairly similar pattern, but with less decay (from 0.84 to 0.70 over the 8 months). The flushed-water Pb correlations are estimated to remain constant at about 0.66. As might be anticipated, lower lag correlations were observed for the VOCs in personal air (due to large intra-person variability relative to the intra-home variability in participants' water supplies). Except for TCE, for which the correlations fluctuated considerably, the VOCs were estimated to have stable correlations over time. The relatively high correlations between longitudinal observations found for Pb and As tap water samples indicate that the within-home (i.e., between-visit) variation is small relative to the between-home variation and suggest that only one or two visits per HH may provide a reasonably good long-term estimate for a given HH. For VOCs in personal air, the lower longitudinal correlations imply that the within-person variation was larger relative to the between-person variation, and suggest that more than one or two 6-day observations per person may often be needed to accurately estimate an individual's long-term exposure level.

One of the goals of the longitudinal component of the study was to explore the possibility of using model-based approaches to estimate distributions of long-term (e.g.,

Table 10. Correlations of longitudinal observations, by medium, analyte, and lag time.

Medium	Analyte	Statistic*	Lag time (months) between observations							
			1	2	3	4	5	6	7	8
Standing water	As	*n*	40	59	50	33	33	22	14	23
		L. Corr	0.95	0.90	0.83	0.74	0.55	0.43	0.19	0.72
		Est.	0.92	0.85	0.78	0.70	0.64	0.57	0.51	0.46
Flushed water	As	*n*	42	60	51	32	33	22	14	23
		L. Corr	0.95	0.92	0.84	0.71	0.55	0.69	0.31	0.72
		Est.	0.93	0.86	0.80	0.74	0.68	0.63	0.59	0.54
Standing water	Pb	*n*	40	58	51	33	33	22	14	22
		L. Corr	0.84	0.80	0.77	0.45	0.70	0.83	0.84	0.70
		Est.	0.84	0.77	0.73	0.71	0.70	0.70	0.70	0.70
Flushed water	Pb	*n*	42	60	50	32	33	20	14	22
		L. Corr	0.42	0.55	0.78	0.82	0.70	0.70	0.80	0.68
		Est.	0.66	0.66	0.66	0.66	0.66	0.66	0.66	0.66
Personal air	Benzene	*n*	33	43	34	24	17	12	17	12
		L. Corr	0.36	0.42	0.43	0.62	0.72	0.46	0.41	0.65
		Est.	0.48	0.48	0.48	0.48	0.48	0.48	0.48	0.48
Personal air	Chloroform	*n*	33	43	34	23	17	12	17	12
		L. Corr	0.11	0.40	0.29	0.51	0.37	0.28	0.08	−0.37
		Est.	0.26	0.26	0.26	0.26	0.26	0.26	0.26	0.26
Personal air	PERC	*n*	33	43	34	24	17	12	17	12
		L. Corr	0.35	0.64	0.46	0.76	0.54	0.56	0.75	−0.01
		Est.	0.53	0.53	0.53	0.53	0.53	0.53	0.53	0.53
Personal air	TCE	*n*	33	43	34	24	17	12	17	12
		L. Corr	0.28	0.73	0.32	0.78	−0.09	0.53	0.17	−0.10
		Est.	0.72	0.53	0.41	0.33	0.28	0.24	0.22	0.21

*n=number of pairs involved in correlation.

L. Corr=correlation (unweighted Pearson) between (log-scale) observations for the same participant.

Est.=a smoothed estimate of the correlation based on the model $\hat{\rho}_k = a + (1-a)e^{-bk}$, where k denotes the lag time and a and b are estimated parameters.

annual) exposures. A possible approach is an adaptation of the methodology of Wallace et al. (1994). This approach assumes that the population distribution of exposures for a 6-day monitoring period, say, can be approximated with a lognormal distribution; it further assumes that the distribution of averages of exposures over two such 6-day periods, or the distribution of averages over any number of 6-day periods can be similarly approximated. If the log-scale variances of these various lognormal distributions are assumed to be comprised of a long-term component of variation (denoted by γ_L) and a short-term component (denoted by γ_S), then Wallace et al. show that these parameters can be expressed as

$$V_T = \gamma_L + \log\left[1 + \frac{e^{\gamma_s} - 1}{T}\right]$$

where T denotes the number of periods over which the average is taken and V_T denotes the log-scale variance for the lognormal distribution associated with T periods. If estimates of V_T can be determined for at least two values of T (e.g., $T=1$ and $T=2$ if participants have two visits, or $T=1$ and $T=3$ if participants have three visits), then these estimates can be substituted for the V_T in the above, yielding two equations that can be simultaneously solved to produce estimates of the short- and long-term variance components. From these estimates, the lognormal parameters for any T can be estimated by substituting into the equation above; this includes $T=61$, for instance, which corresponds to an annual period (i.e., 61 six-day periods). The basic notion of the method is that, while the mean of the exposure distributions stays constant, the variability decreases as the number of periods averaged together increases. Hence, geometric standard deviations (GSDs) are expected to decrease and geometric means are expected to increase as T increases. The above described method represents a modification of the original procedure in two ways. First, it is based on estimated population variances (from a weighted data analysis), and second, it assumes that the goal is an estimated annual (rather than a "lifetime") distribution.

Checks of the lognormality assumptions underlying the procedure were first made by constructing probability plots of the weighted empirical distribution functions. These plots were done for each visit separately and for averages over the first two visits and over all three visits. These plots were examined and the results are summarized in Table 11. The As and VOC cases did not appear to meet the assumptions; as a result, the approach was applied only to the two Pb cases (standing and flushed tap water). (For the water samples, which were taken at a single point in time during the 6-day monitoring period, one might argue that the sample data points do not represent the entire 6-day period;

Table 11. Assessment of lognormality of population distributions, based on probability plots of the weighted empirical distribution function.[a]

Medium	Analyte	Visit 1 data	Visit 2 data	Visit 3 data	Visits 1, 2, 3 combined	Avg. of 2 visits	Avg. of 3 visits
Standing	As	F	P	P	P	P	P
Water	Pb	G	G	VG	G	VG	VG
Flushed	As	P	P	P	P	P	P
Water	Pb	F	VG	VG	G	G	G
Personal	Benzene	P	P	P	P	P	F
Air	Chloroform	F	F	P	F	F	G
	PERC	P	P	P	P	P	P
	TCE	P	G	P	F	F	G

[a]Adequacy of the lognormal approximation was rated using the following notation: P=poor, F=fair, G=good, VG=very good.

for purposes of demonstrating the procedure, we make that assumption. One might also be able to find population subgroups for which the lognormal assumptions might well hold for some of the other cases. For instance, if only nonsmokers were considered, then the benzene personal air distributions might be substantially different from the overall population distributions and those distributions might be well approximated by lognormal distributions.)

Table 12 presents the results of applying the procedure to the Pb standing-water concentrations. The following four ways (cases) of applying the procedure were utilized.

Case I: Estimates of the log-scale parameters were obtained (using visit-specific sampling weights) for each visit and for the two-visit averages. The single-visit estimates were weighted by sample size to produce a single log-scale mean and standard deviation (utilizing 444 observations). These were exponentiated to produce the geometric mean and standard deviation at the top of the table. The short- and long-term variance components were then determined. For this case, a negative short-term component was reset to zero. The resultant distribution is thus static (i.e., does not depend on averaging time).

Case II: Estimates of the log-scale parameters were obtained for each visit and for the three-visit averages. The single-visit estimates were treated in the same way as for Case I. In this case, which is probably preferable to Case I since it makes use of more information, the GSDs and the geometric means exhibit the expected patterns as T increases. The lower portion of the table shows how the 90th percentiles are estimated to change with increasing T.

Case I-A: Since log-scale means and variances for lognormal distributions can be derived from known relationships to the mean and coefficient of variation (CV) of the distribution, estimates of the mean and CV (on the concentration scale) were first determined for each visit

Table 12. Estimates of short- and long-term variance components and resultant long-term distributions based on an adaptation of the Wallace et al. methodology—for Pb in standing water.

Statistic	Case I	Case II	Case I-A	Case II-A
One period				
N	444	444	444	444
Geometric mean	1.89	1.89	2.23	2.23
GSD	3.56	3.56	2.89	2.89
Two-period average				
N	105		105	
Geometric mean	2.24		2.94	
GSD	3.72		2.64	
Three-period average				
N		88		88
Geometric mean		1.91		2.28
GSD		3.08		2.41
Estimated variance components				
Short-term	0.000	0.567	0.411	0.593
Long-term	1.666	1.042	0.714	0.532
Estimated geometric mean for:				
T=1	1.84	1.89	2.23	2.23
T=2	1.84	2.14	2.45	2.54
T=3	1.84	2.24	2.54	2.67
T=61	1.84	2.50	2.73	2.98
Estimated GSD for:				
T=1	3.64	3.56	2.89	2.89
T=2	3.64	3.22	2.64	2.54
T=3	3.64	3.08	2.54	2.41
T=61	3.64	2.79	2.34	2.09
Estimated 90th percentile for:				
T=1	15.4	15.2	12.8	12.8
T=2	15.4	14.6	12.1	11.8
T=3	15.4	14.3	11.8	11.3
T=61	15.4	13.5	11.1	10.1

Case I: Variance components based on direct estimates of log-scale variances for single visits (combined) and two-visit averages.
Case II: Variance components based on direct estimates of log-scale variances for single visits (combined) and three-visit averages.
Case I-A: Like Case I, but using alternate estimates of log-scale variances derived from estimated coefficients of variation of the concentration-scale data.
Case II-A: Like Case II, but using alternate estimates of log-scale variances derived from estimated coefficients of variation of the concentration-scale data.

and for the two-visit average. The corresponding log-scale parameters were then computed. Except for using these alternative estimates of the log-scale parameters, Case I-A is like Case I. This approach led to lower estimates for the 90th percentiles.

Table 13. Estimates of short- and long-term variance components and resultant long-term distributions based on an adaptation of the Wallace et al. methodology—for Pb in flushed water.

Statistic	Case I	Case II	Case I-A	Case II-A
One period				
N	443	443	443	443
Geometric mean	0.32	0.32	0.32	0.32
GSD	3.82	3.82	4.01	4.01
Two-period average				
N	105		105	
Geometric mean	0.41		0.41	
GSD	3.16		3.14	
Three-period average				
N		87		87
Geometric mean		0.39		0.46
GSD		3.43		2.90
Estimated variance components				
Short-term	1.392	0.460	1.894	1.740
Long-term	0.406	1.338	0.000	0.189
Estimated geometric mean for:				
T=1	0.32	0.32	0.33	0.32
T=2	0.40	0.35	0.43	0.42
T=3	0.45	0.37	0.50	0.48
T=61	0.62	0.40	0.81	0.74
Estimated GSD for:				
T=1	3.82	3.82	3.96	4.01
T=2	3.16	3.53	3.18	3.26
T=3	2.86	3.43	2.80	2.90
T=61	1.96	3.19	1.35	1.67
Estimated 90th percentile for:				
T=1	1.78	1.78	1.92	1.92
T=2	1.77	1.78	1.92	1.92
T=3	1.74	1.78	1.87	1.89
T=61	1.48	1.77	1.19	1.44

Case I: Variance components based on direct estimates of log-scale variances for single visits (combined) and two-visit averages.
Case II: Variance components based on direct estimates of log-scale variances for single visits (combined) and three-visit averages.
Case I-A: Like Case I, but using alternate estimates of log-scale variances derived from estimated coefficients of variation of the concentration-scale data.
Case II-A: Like Case II, but using alternate estimates of log-scale variances derived from estimated coefficients of variation of the concentration-scale data.

Case II-A: Estimates of the mean and CV were determined for each visit and for the three-visit average and log-scale parameter estimates were then computed to be consistent with these estimates in the manner described for Case I-A. Except for using these alternative estimates of the log-scale parameters, Case II-A is like Case II. This approach led to even lower estimates for the 90th percentiles.

It should be noted that if the lognormal assumptions and the assumed model are accurate and if the data are adequate to produce accurate estimates for the GSDs and geometric means, then each of the above approaches should produce similar annual distributions. This appears not to be the case, however.

Table 13 shows comparable results for the flushed water concentrations. Again, the various cases yield somewhat disparate results.

Conclusions

The distributional results and associations provided herein should provide a useful starting point for understanding the relative importance of different routes of human exposures to As, Pb, and VOCs. In particular, it appears that solid food represents an important component of exposure for As, as evidenced by significant correlations between urine levels and solid food concentrations and intakes. On the other hand, for Pb, several routes appear to be important, as suggested by the high dust and beverage levels and the significant correlations of dust, water, personal air, and beverage concentrations with blood Pb levels.

Statistically significant correlations were found for blood VOC levels versus personal air VOC levels; some of the VOC distributions exhibited extreme skewness, suggesting the possibility of occupational exposures for some participants.

Relatively high correlations between longitudinal observations were found for Pb and As tap water samples, suggesting that temporal (i.e., visit-within-home) variation is small relative to the between-home variation and suggesting that only one or two visits per HH will provide a good long-term estimate for a given HH for these analytes in water. VOCs in personal air exhibited lower longitudinal correlations, implying a somewhat larger visit-to-visit variation relative to between-person variation, and suggesting that more than one or two 6-day observations per person may often be needed to estimate accurately an individual's long-term exposure level. Attempts to estimate long-term distributions of exposures were hampered by failure of most media/analyte combinations to satisfy the underlying lognormality assumptions; for those combinations where the assumptions did appear valid, the procedure did not appear to produce consistent results over several alternative estimation strategies. Further consideration of such models and methods (e.g., defining relevant population subgroups for which the assumptions are more tenable or developing methods with less restrictive assumptions) is needed.

Acknowledgments

We thank K. Thomas, L. Cauble, L. Klemm, M. Herrmann, and J. Roberds for their expert conduct of field activities and L. Michael for his development of the study database. We also thank the many participants who provided the data. This research was supported by EPA Cooperative Agreement CR821902-01-0.

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